

FORM PTO-1390 (REV 3-93)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE ATTORNEY'S DOCKET NUMBER	
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		von Kreisler.021	
		U.S. APPLICATION NO. (if known, see 37 CFR 1.5) <b>10/069476</b>	
INTERNATIONAL APPLICATION NO. PCT/EP00/08344	INTERNATIONAL FILING DATE 25/08/00	PRIORITY DATE CLAIMED 25/08/99	
TITLE OF INVENTION "PLANT PROTECTION"			
APPLICANT(S) FOR DO/EO/US Jörg Peter Schür			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<ol style="list-style-type: none"> <li>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</li> <li>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</li> <li>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)</li> </ol> </li> <li>6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</li> <li>7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> <li>a. <input checked="" type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> have been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li>8. <input checked="" type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</li> <li>9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</li> <li>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</li> </ol>			
Items 11. to 16. below concern other document(s) or information included:			
11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.			
12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.			
13. <input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment. <input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.			
14. <input checked="" type="checkbox"/> A substitute specification.			
15. <input type="checkbox"/> A change of power of attorney and/or address letter.			
16. <input type="checkbox"/> Other items or information:			

10069476.070102

10/069476

JC19 Rec'd PCT/PTO 25 FEB 2002

PTO/SB/17 (11-01)

Approved for use through 10/31/2002. OMB 0651-0032  
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

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**FEE TRANSMITTAL  
for FY 2002**

Patent fees are subject to annual revision.

☒ Applicant claims small entity status. See 37 CFR 1.27

TOTAL AMOUNT OF PAYMENT

(\$) 658.00

**Complete if Known**

Application Number

Filing Date

First Named Inventor

Jörg Peter Schür

Examiner Name

Group Art Unit

Attorney Docket No.

von Kreisler.021

**METHOD OF PAYMENT (check all that apply)**☒ Check ☐ Credit card ☐ Money Order ☐ Other ☐ None☒ Deposit Account:Deposit  
Account  
Number  
Deposit  
Account  
Name

04-1406

Dann Dorfman Herrell  
and Skillman

The Commissioner is authorized to: (check all that apply)

☐ Charge fee(s) indicated below ☒ Credit any overpayments☒ Charge any additional fee(s) during the pendency of this application☐ Charge fee(s) indicated below, except for the filing fee  
to the above identified deposit account.**FEE CALCULATION****1. BASIC FILING FEE**

Large Entity Fee Code (\$)	Small Entity Fee Code (\$)	Fee Description	Fee Paid
101 740	201 370	Utility filing fee	370.00
106 330	206 165	Design filing fee	
107 510	207 255	Plant filing fee	
108 740	208 370	Reissue filing fee	
114 160	214 80	Provisional filing fee	

SUBTOTAL (1) (\$) 370.00

**2. EXTRA CLAIM FEES FOR UTILITY AND REISSUE**

Extra Claims Fee from below Fee Paid

Total Claims 52 -20\*\* = 32 x 9.00 = 288.00

Independent Claims 3 -3\*\* = 0 x 42.00 = 0

Multiple Dependent 0 x 140.00 = 0

Large Entity Fee Code (\$)	Small Entity Fee Code (\$)	Fee Description
103 18	203 9	Claims in excess of 20
102 84	202 42	Independent claims in excess of 3
104 280	204 140	Multiple dependent claim, if not paid
109 84	209 42	** Reissue independent claims over original patent
110 18	210 9	** Reissue claims in excess of 20 and over original patent

SUBTOTAL (2) (\$) 288.00

\*\*or number previously paid, if greater; For Reissues, see above

**FEE CALCULATION (continued)****3. ADDITIONAL FEES**

Large Entity Small Entity

Fee Code (\$)	Fee Code (\$)	Fee Description	Fee Paid
105 130	205 65	Surcharge - late filing fee or oath	
127 50	227 25	Surcharge - late provisional filing fee or cover sheet	
139 130	139 130	Non-English specification	
147 2,520	147 2,520	For filing a request for <i>ex parte</i> reexamination	
112 920*	112 920*	Requesting publication of SIR prior to Examiner action	
113 1,840*	113 1,840*	Requesting publication of SIR after Examiner action	
115 110	215 55	Extension for reply within first month	
116 400	216 200	Extension for reply within second month	
117 920	217 460	Extension for reply within third month	
118 1,440	218 720	Extension for reply within fourth month	
128 1,960	228 980	Extension for reply within fifth month	
119 320	219 160	Notice of Appeal	
120 320	220 160	Filing a brief in support of an appeal	
121 280	221 140	Request for oral hearing	
138 1,510	138 1,510	Petition to institute a public use proceeding	
140 110	240 55	Petition to revive - unavoidable	
141 1,280	241 640	Petition to revive - unintentional	
142 1,280	242 640	Utility issue fee (or reissue)	
143 460	243 230	Design issue fee	
144 620	244 310	Plant issue fee	
122 130	122 130	Petitions to the Commissioner	
123 50	123 50	Processing fee under 37 CFR 1.17(q)	
126 180	126 180	Submission of Information Disclosure Stmt	
581 40	581 40	Recording each patent assignment per property (times number of properties)	
146 740	246 370	Filing a submission after final rejection (37 CFR § 1.129(a))	
149 740	249 370	For each additional invention to be examined (37 CFR § 1.129(b))	
179 740	279 370	Request for Continued Examination (RCE)	
169 900	169 900	Request for expedited examination of a design application	

Other fee (specify)

\*Reduced by Basic Filing Fee Paid

SUBTOTAL (3) (\$) 0

**SUBMITTED BY**

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John S. Child, Jr.

Date

25/02/00

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Burden Hour Statement: This form is estimated to take 0.2 hours to complete. Time will vary depending upon the needs of the individual case. Any comments on the amount of time you are required to complete this form should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, Washington, DC 20231. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Assistant Commissioner for Patents, Washington, DC 20231.

10069476 10/069476

JC19 Rec'd PCT/PTO 25 FEB 2002

PATENT

ATTORNEY DOCKET NO. VON KREISLER.021

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Application for National Phase  
Filing in the United States of

Applicant: Jörg Peter Schür

Serial No.: [to be assigned]

Filing Date: [concurrently herewith]

Based upon International Application No.  
PCT/EP00/08344

Filed : August 25, 2000

For: "PLANT PROTECTION"

Examiner:

Group Art Unit:

**BOX — FEE — PCT**

Assistant Commissioner for Patents  
Washington, DC 20231

**PRELIMINARY AMENDMENT UNDER 37 C.F.R. § 1.111**

Dear Sir:

Applicant amends the specification and claims as follows:

**In The Specification:**

Please insert the following paragraph at page 1, line 1:

— —This Application is a national phase filing of co-  
pending International Application No. PCT/EP00/08344  
filed August 25, 2000, which claims the benefit of that  
application under 35 U.S.C. § 120 and which also claims  
the benefit under 35 U.S.C. § 119 of German Application  
No. 119 40 283.3 filed August 25, 1999. — —

**In The Claims:**

Please amend the claims as follows:

1. (Amended) A method for protecting plants shortly before or after the harvest from microbial attack, comprising the [distribution or application of] step of applying an antimicrobial composition to the surface of the plants, said antimicrobial composition [containing] comprising:
  - [(ii)] (i) at least one lipophilic GRAS (generally recognized as safe) flavoring agent; and
  - (ii) at least one hydrophilic GRAS flavoring agent.
2. (Amended) The method according to claim 1, wherein said lipophilic GRAS flavoring agents are selected from the group consisting of (a<sub>1</sub>) lipophilic GRAS flavor alcohols or their derivatives, (b) polyphenol compounds, (c<sub>1</sub>) lipophilic GRAS flavor acids or their derivatives, (d) phenols or their derivatives, (e<sub>1</sub>) lipophilic esters, (f) terpenes, (g) acetals, (h<sub>1</sub>) lipophilic aldehydes and (i) essential oils.
3. (Amended) The method according to claim 1 [or 2], wherein said antimicrobial composition [contains] comprises at least two lipophilic GRAS flavoring agents[, preferably two lipophilic GRAS flavor alcohols (a<sub>1</sub>)].

4. (Amended) The method according to claim 2 [or 3], wherein said lipophilic GRAS flavor alcohols are selected from[:] the group consisting of aromatic GRAS flavor alcohols, including benzyl alcohol, 2-phenylethanol, 1-phenylethanol, cinnamyl alcohol, hydrocinnamyl alcohol, 1-phenyl-1-propanol and anisalcohol, and aliphatic GRAS flavor alcohols, including n-butyl alcohol, iso-butyl alcohol, hexyl alcohol, L-menthol, octyl alcohol, heptyl alcohol, n-amyl alcohol, iso-amyl alcohol, anisalcohol, citronellol, n-decyl alcohol, geraniol,  $\beta$ , $\gamma$ -hexenol, lauryl alcohol, linalool, nerolidol, nonadienol, nonyl alcohol, rhodinol, terpeneol, borneol, clineol, anisole, cuminyl alcohol, 10-undecene-1-ol and 1-hexadecanol and their derivatives[, said aromatic GRAS flavor alcohols, especially benzyl alcohol, being preferred].
5. (Amended) The method according to [one or more of claims 1 to 4] claim 2, wherein said hydrophilic GRAS flavoring agent is selected from the group consisting of a hydrophilic alcoholic GRAS flavoring agent [(a<sub>n</sub>) or] and a hydrophilic non-alcoholic GRAS flavoring agent, wherein said [hydrophilic alcoholic GRAS flavoring agent (a<sub>n</sub>) is preferably a monohydric or polyhydric alcohol having from 2 to 10, more preferably from 2 to 7, carbon atoms, especially one selected from acetoin, ethyl alcohol, propyl alcohol, isopropyl alcohol, propylene glycol and glycerol, and said] hydrophilic non-alcoholic GRAS flavoring agent is a hydrophilic organic GRAS flavor acid [(c<sub>n</sub>)] having from 1 to

15 carbon atoms or a physiological salt thereof, a hydrophilic acetate [(e<sub>h</sub>)] or a hydrophilic aldehyde [(h<sub>h</sub>)].

6. (Amended) The method according to claim 5, wherein said hydrophilic organic acid [(c<sub>h</sub>)] has from 2 to 10 carbon atoms[, especially being selected from acetic acid, aconitic acid, formic acid, malic acid, lactic acid, phenylacetic acid, citric acid, mandelic acid, tartaric acid, fumaric acid, tannic acid, hydrocinnamic acid and their physiological salts;] said hydrophilic acetate [(e<sub>h</sub>)] is selected from the group consisting of allucin, triacetin, potassium acetate, sodium acetate and calcium acetate; and[/or] said hydrophilic aldehyde [(h<sub>h</sub>)] is selected from the group consisting of furfural, propionaldehyde and vanillin.
7. (Amended) The method according to claim 5, wherein said antimicrobial composition [contains] comprises less than 50% by weight[, preferably less than 30% by weight, more preferably less than 20% by weight,] of benzyl alcohol or of a mixture of benzyl alcohol with ethanol and/or isopropanol.
8. (Amended) The method according to claim 5 [or 6], wherein said antimicrobial composition [contains] comprises two lipophilic GRAS flavor alcohols [(a<sub>i</sub>)], but no benzyl alcohol and no polyphenol compounds [(b)].

9. (Amended) The method according to claim 5 [or 6], wherein said antimicrobial composition [contains] comprises benzyl alcohol and/or a polyphenol compound [(b)], but no further GRAS flavor alcohols.
10. (Amended) The method according to claim 8 [or 9], wherein said antimicrobial composition [exclusively contains non-alcoholic hydrophilic GRAS flavoring agents, especially exclusively] consists of a hydrophilic GRAS flavor acid [(c<sub>h</sub>)].
11. (Amended) The method according to claim 9 [or 10], wherein said antimicrobial composition [contains] comprises from 0.01 to 99% by weight[, preferably from 0.1 to 90% by weight,] of benzyl alcohol or polyphenol compounds [(b)] and from 0.01 to 50% by weight[, preferably from 0.1 to 30% by weight,] of hydrophilic non-alcoholic GRAS flavoring agents.
12. (Amended) The method according to claim 1 [or 2], wherein said antimicrobial composition [contains] comprises:
  - (A) at least one [or more] GRAS flavor alcohol[s] (a) and/or [their] its derivatives; and
  - (B) at least one [or more] flavoring agent[s] selected from the group consisting of polyphenol compounds (b) and lipophilic GRAS flavor acids or their derivatives (c).

13. (Amended) The method according to claim 12, wherein said antimicrobial composition [contains] comprises:  
  
 from 0.1 to 99% by weight[, preferably from 0.5 to 99% by weight,] of component (a),  
 from 0 to 25% by weight[, preferably from 0.01 to 10% by weight,] of component (b), and  
 from 0 to 70% by weight[, preferably from 0.01 to 30% by weight,] of component (c).
14. (Amended) The method according to claim 12 [or 13], wherein said antimicrobial composition contains further GRAS flavoring agents selected from (d) phenols or their derivatives, (e<sub>i</sub>) lipophilic esters, (f) terpenes, (g) acetals, (h<sub>i</sub>) lipophilic aldehydes and (i) essential oils.
15. (Amended) The method according to [one or more of] claim[s] 12 [to 14], wherein component (A) of said antimicrobial composition [contains] is benzyl alcohol [as a necessary component (a<sub>1</sub>) and optionally one or more further lipophilic GRAS flavor alcohols or their derivatives (a<sub>i</sub>)].
16. (Amended) The method according to [one or more of] claim[s] 12 to 15] 13, wherein said polyphenol compound [(b)] is selected from the group consisting of:



catechol, resorcinol, hydroquinone, phloroglucinol, pyrogallol, cyclohexane, resveratrol, usnic acid, acylpolyphenols, lignins, anthocyanins, flavones, catechols, gallic acid derivatives, caffeic acid, flavonoids, derivatives of the mentioned polyphenols, and extracts from Camellia, Primula; and said lipophilic GRAS acid [(c)] is selected from the group consisting of:  
adipic acid, capronic acid, pelargonic acid, phenoxyacetic acid, valeric acid, iso-valeric acid, cinnamic acid, mandelic acid and their derivatives.

17. (Amended) The method according to claim [15 or 16] 12, wherein the component A of said antimicrobial composition [contains] comprises:

from 0.1 to 99% by weight[, preferably from 0.1 to 75% by weight,] of benzyl alcohol;

from 0 to 99.8% by weight[, preferably from 0.01 to 99% by weight,] of component (a); [and]

from 0 to 25% by weight[, preferably from 0.01 to 10% by weight,] of component (b); and

from 0 to 70% by weight[, preferably from 0.01 to 30% by weight,] of component (c).

18. (Amended) The method according to claim 17, wherein said antimicrobial composition [contains] comprises from 0.001 to 25% by

weight of further lipophilic GRAS flavoring agents [(d) to (i), preferably from 0.001 to 25% by weight, more preferably from 0.01 to 9% by weight, of said further GRAS flavoring agents [(d) to (i)] selected from the group consisting of phenols or their derivatives, esters, terpenes, acetals, aldehydes and essential oils or extracts thereof.

19. *(Amended)* The method according to claim 18, wherein said further lipophilic GRAS flavoring agents are phenols [(d)] and/or essential oils [(i)] or extracts thereof having a high content of alcohols, aldehydes, phenols, acetates or esters.
20. *(Amended)* The method according to [one or more of] claim[s] 1 [to 19], wherein said antimicrobial composition does not contain any derivatives of the GRAS flavoring agents.
21. *(Amended)* The method according to [one or more of] claim[s] 15 to 20] 17, wherein said antimicrobial composition [contains] comprises one or two lipophilic GRAS flavor alcohols [(a<sub>1</sub>)] and at least one polyphenol compound [(b)].
22. *(Amended)* The method according to claim 21, wherein said polyphenol compound [(b)] is tannin.

24. (Amended) The method according to [one or more of] claim[s] 1 [to 23], wherein said antimicrobial composition further [contains] comprises monohydric or polyhydric alcohols having from 2 to 10 carbon atoms, and additives selected from the group consisting of emulsifiers, stabilizers, antioxidants, preservatives, solvents and[/or] carriers.
25. (Amended) The method according to [one or more of] claim[s] 1 [to 23], wherein said antimicrobial composition [exclusively] consists of GRAS flavoring agents.
26. (Amended) The method according to [one or more of] claim[s] 1 [to 25], wherein said [distribution or application] step of applying [comprises application of] said antimicrobial composition to the surface of the plants[, especially] is carried out by spraying, immersion or nebulizing.
27. (Amended) A method for protecting plants from microbial attack shortly before or after the harvest, comprising the distribution of said antimicrobial composition within the plant[, which is effected] by the step of adding the antimicrobial composition to nutrient media, nutrient liquids and/or water, said antimicrobial composition [containing] comprising at least two GRAS (generally recognized as safe) flavoring agents.

28. (Amended) The method according to claim 27, wherein said GRAS flavoring agents are selected from the group consisting of (a) GRAS flavor alcohols or their derivatives, (b) polyphenol compounds, (c) GRAS flavor acids or their derivatives, (d) phenols or their derivatives, (e) esters, (f) terpenes, (g) acetals, (h) aldehydes and (i) essential oils.
29. (Amended) The method according to claim 27 [or 28], wherein said antimicrobial composition [contains] comprises at least one GRAS flavor alcohol [(a), preferably an aromatic GRAS flavor alcohol, especially benzyl alcohol].
30. (Amended) The method according to claim 29, wherein said antimicrobial composition [contains] comprises less than 50% by weight[, preferably less than 30% by weight, more preferably less than 20% by weight,] of an alcohol selected from the group consisting of ethanol, isopropanol or benzyl alcohol or a mixture of these [substances] alcohols.
31. (Amended) The method according to claim 27 [or 28], wherein said antimicrobial composition [contains] comprises at least one hydrophilic alcoholic GRAS flavoring agent and/or one hydrophilic non-alcoholic GRAS flavoring agent.

32. (Amended) The method according to claim 31, wherein said antimicrobial composition further [contains] comprises benzyl alcohol and/or a polyphenol compound [(b)].
33. (Amended) The method according to claim 27 [or 28], wherein said antimicrobial composition [contains] comprises:
- (A) at least one [or more] GRAS flavor alcohol[s] (a) and/or [their] its derivative[s]; and
- (B) at least one [or more] flavoring agent[s] selected from the group consisting of (b) polyphenol compounds[:]; and (c) GRAS flavor acids or their derivatives.
34. (Amended) The method according to claim 33, wherein said antimicrobial composition [contains] comprises:
- from 0.1 to 99% by weight[, preferably from 0.5 to 99% by weight,] of [component (a),] GRAS flavor alcohol;
- from 0 to 25% by weight[, preferably from 0.01 to 10% by weight,] of polyphenol compounds; [component (b),] and
- from 0 to 70% by weight[, preferably from 0.01 to 30% by weight,] of [component (c)] GRAS flavor acids or their derivatives.
35. (Amended) The method according to [one or more of] claim[s] 27 [to 34], wherein said antimicrobial composition [contains] comprises benzyl alcohol and at least one further GRAS flavoring agent.

37. (Amended) The method according to claim 36, wherein said antimicrobial composition [contains] comprises less than 50% by weight[, preferably less than 30% by weight, more preferably less than 20% by weight,] of benzyl alcohol or of a mixture of benzyl alcohol with ethanol and/or isopropanol.
38. (Amended) The method according to [one or more of] claim[s] 33 [to 36], wherein said antimicrobial composition [contains] comprises:
- [(a1)] benzyl alcohol as a necessary component; [and optionally
- (a2)] at least one [or more further] other GRAS flavor alcohol[s] and/ or their derivatives; [and]
- [(b)] at least one [or more] polyphenol compound[s]; and/or
- [(c)] at least one [or more] GRAS acid[s] and/ or their derivatives.
39. (Amended) The method according to claim 38, wherein said further GRAS flavor alcohol [(a2)] is selected from the group consisting of: acetoin, ethyl alcohol, propyl alcohol, isopropyl alcohol, propylene glycol, glycerol, n-butyl alcohol, iso-butyl alcohol, hexyl alcohol, L-menthol, octyl alcohol, cinnamyl alcohol,  $\alpha$ -methylbenzyl alcohol, heptyl alcohol, n-amyl alcohol, iso-amyl alcohol, anisalcohol, citronellol, n-decyl alcohol, geraniol,  $\beta,\gamma$ -hexenol, lauryl alcohol, linalool, nerolidol, nonadienol, nonyl alcohol, rhodinol, terpineol, borneol, clineol, anisole, cuminyl alcohol, 10-un-decene-1-ol, 1-hexadecanol or their derivatives;

said polyphenol compound [(b)] is selected from the group consisting of:

catechol, resorcinol, hydroquinone, phloroglucinol, pyrogallol, cyclohexane, resveratrol, usnic acid, acylpolyphenols, lignins, anthocyanins, flavones, catechols, gallic acid derivatives, caffeic acid, flavonoids, derivatives of the mentioned polyphenols, and extracts from Camellia, Primula; and

said GRAS acid [(c)] is selected from the group consisting of:

acetic acid, aconitic acid, adipic acid, formic acid, malic acid, capronic acid, hydrocinnamic acid, pelargonic acid, lactic acid, phenoxyacetic acid, phenylacetic acid, valeric acid, iso-valeric acid, cinnamic acid, citric acid, mandelic acid, tartaric acid, fumaric acid, tannic acid and their derivatives.

40. (Amended) The method according to claim 38 [or 39], wherein said antimicrobial composition [contains] comprises:

from 0.1 to 99% by weight[, preferably from 0.1 to 75% by weight,] of benzyl alcohol;

from 0 to 99.8% by weight[, preferably from 0.01 to 99% by weight,] of other GRAS flavor alcohols and/or their derivatives [component (a2)];

[and]

from 0 to 25% by weight[, preferably from 0.01 to 10% by weight,] of  
polyphenol compounds [component (b)]; and  
from 0 to 70% by weight[, preferably from 0.01 to 30% by weight,] of  
[component (c)] GRAS acids and/or their derivatives.

41. (Amended) The method according to [one or more of] claim[s] 38 [to 40], wherein said antimicrobial composition [contains] comprises further GRAS flavoring agents selected from the group consisting of [(d)] phenols, [(e)] esters, [(f)] terpenes, [(g)] acetals, [(h)] aldehydes and [(i)] essential oils.
42. (Amended) The method according to claim 41, wherein said antimicrobial composition contains from 0.001 to 25% by weight[, preferably from 0.01 to 9% by weight,] of said further GRAS flavoring agents [(d) to (i)].
43. (Amended) The method according to claim [41 or] 42, wherein said further GRAS flavoring agents are phenols [(d)] and/or essential oils [(i)].
44. (Amended) The method according to [one or more of] claim[s] 27 [to 43], wherein said antimicrobial composition does not contain any derivatives of the GRAS flavoring agents.



45. (Amended) The method according to [one or more of] claim[s] 38 [to 44], wherein said antimicrobial composition contains one or two GRAS flavor alcohols [(a2)] and at least one polyphenol compound [(b)].
46. (Amended) The method according to claim 45, wherein said polyphenol compound [(b)] is tannin.
48. (Amended) The method according to claim 27, wherein said antimicrobial composition is the composition of [defined as in claims 1 to 25, especially as in] claim[s] 12 [to 25].
49. (Amended) A method for protecting plants shortly before or after the harvest from insects and insect larvae, comprising the steps of:
  - (i) [the distribution or application of] applying an insecticidal composition to the surface of the plants and/or
  - (ii) [the distribution of] distributing an insecticidal composition within the plant by adding the insecticidal composition to nutrient media, nutrient liquids and/or water; wherein said insecticidal composition is a composition containing GRAS flavoring agents as defined in claim[s] 1 [to 25 and 27 to 48].

50. (Amended) The method according to claim 49, wherein the step of applying [said distribution or application of] the insecticidal composition to the surface of the plant [comprises application of said insecticidal composition to the surface of the plants, especially] is carried out by spraying, immersion or nebulizing.
51. (Amended) The method according to [one or more of] claim[s] 1 [to 50], wherein said plants are selected from the group consisting of cotton, cereals, rice, corn, potatoes, tobacco, coffee, cocoa, tea, vegetables, fruits, nuts, spices, herbs, seeds, ornamental plants, cultured flowers and flowers for cutting.
52. (Amended) The method according to claim 1, wherein the [use of an] antimicrobial composition as defined in claim[s] 1 [to 25 and 27 to 48 for the treatment of plants] is applied shortly before and after the harvest to prevent molds, viruses and parasites.

### **REMARKS**

The specification has been amended to claim the benefits of the copending International Application under 35 U.S.C. § 120 and of the priority German patent application under 35 U.S.C. § 119.

The translation of the International Application did not include an Abstract of the Disclosure as required under 37 C.F.R. § 1.72(b). However, the copending

International Application, when published as Published International Application No. WO 01/13727A1, on March 1, 2001, included an Abstract in English. That Abstract is included in the English language copy of the application submitted herewith and is considered to be part of the application because of its inclusion in the Published International Application No. WO 01/13727A1.

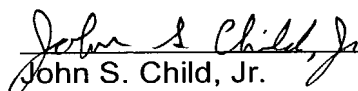
Certain of pending claims 1–52 have been amended to remove multiple dependencies and try to place the claims in proper form under United States Patent and Trademark Office practice. There is clear support for the amended claims 1-51 in the corresponding original claims.

Amended Claim 52 has been changed from a use claim to a method of treatment. Support for the additional subject matter in amended claim 52 compared with original claim 52 is provided by the tables at the end of the specification.

A clean copy of the new paragraph in the specification and of claims 1-52 is attached as **Exhibit A** to this Amendment.

Respectfully submitted,

Dann Dorfman Herrell and Skillman  
A Professional Corporation

  
John S. Child, Jr.  
PTO Registration No. 28,833

February 25, 2001

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**PATENT**

ATTORNEY DOCKET NO. VON KREISLER.021

**EXHIBIT A****Amendment to the Specification**

This Application is a national phase filing of co-pending International Application No. PCT/EP00/08344 filed August 25, 2000, which claims the benefit of that application under 35 U.S.C. § 120 and which also claims the benefit under 35 U.S.C. § 119 of German Application No. 119 40 283.3 filed August 25, 1999.

**Claims as Amended**

1. A method for protecting plants shortly before or after the harvest from microbial attack, comprising the step of applying an antimicrobial composition to the surface of the plants, said antimicrobial composition comprising:
  - (i) at least one lipophilic GRAS (generally recognized as safe) flavoring agent; and
  - (ii) at least one hydrophilic GRAS flavoring agent.
2. The method according to claim 1, wherein said lipophilic GRAS flavoring agents are selected from the group consisting of (a) lipophilic GRAS flavor alcohols or their derivatives, (b) polyphenol compounds, (c) lipophilic GRAS flavor acids or their derivatives, (d) phenols or their

derivatives, (e) lipophilic esters, (f) terpenes, (g) acetals, (h) lipophilic aldehydes and (i) essential oils.

3. The method according to claim 1, wherein said antimicrobial composition comprises at least two lipophilic GRAS flavoring agents.
4. The method according to claim 2, wherein said lipophilic GRAS flavor alcohols are selected from the group consisting of aromatic GRAS flavor alcohols, including benzyl alcohol, 2-phenylethanol, 1-phenylethanol, cinnamyl alcohol, hydrocinnamyl alcohol, 1-phenyl-1-propanol and anisalcohol, and aliphatic GRAS flavor alcohols, including n-butyl alcohol, iso-butyl alcohol, hexyl alcohol, L-menthol, octyl alcohol, heptyl alcohol, n-amyl alcohol, iso-amyl alcohol, anisalcohol, citronellol, n-decyl alcohol, geraniol,  $\beta$ , $\gamma$ -hexenol, lauryl alcohol, linalool, nerolidol, nonadienol, nonyl alcohol, rhodinol, terpeneol, borneol, clineol, anisole, cuminyl alcohol, 10-undecene-1-ol and 1-hexadecanol and their derivatives.
5. The method according to claim 2, wherein said hydrophilic GRAS flavoring agent is selected from the group consisting of a hydrophilic alcoholic GRAS flavoring agent and a hydrophilic non-alcoholic GRAS flavoring agent, wherein said hydrophilic non-alcoholic GRAS flavoring agent is a hydrophilic organic GRAS flavor acid having from 1 to 15

carbon atoms or a physiological salt thereof, a hydrophilic acetate or a hydrophilic aldehyde.

6. The method according to claim 5, wherein said hydrophilic organic acid has from 2 to 10 carbon atoms said hydrophilic acetate is selected from the group consisting of allicin, triacetin, potassium acetate, sodium acetate and calcium acetate; and said hydrophilic aldehyde is selected from the group consisting of furfural, propionaldehyde and vanillin.
7. The method according to claim 5, wherein said antimicrobial composition comprises less than 50% by weight of benzyl alcohol or of a mixture of benzyl alcohol with ethanol and/or isopropanol.
8. The method according to claim 5, wherein said antimicrobial composition comprises two lipophilic GRAS flavor alcohols, but no benzyl alcohol and no polyphenol compounds.
9. The method according to claim 5, wherein said antimicrobial composition comprises benzyl alcohol and/or a polyphenol compound, but no further GRAS flavor alcohols.
10. The method according to claim 8, wherein said antimicrobial composition consists of a hydrophilic GRAS flavor acid.

11. The method according to claim 9, wherein said antimicrobial composition comprises from 0.01 to 99% by weight of benzyl alcohol or polyphenol compounds and from 0.01 to 50% by weight of hydrophilic non-alcoholic GRAS flavoring agents.
12. The method according to claim 1, wherein said antimicrobial composition comprises:
  - (A) at least one GRAS flavor alcohol (a) and/or its derivatives; and
  - (B) at least one flavoring agent selected from the group consisting of polyphenol compounds (b) and lipophilic GRAS flavor acids or their derivatives (c).
13. The method according to claim 12, wherein said antimicrobial composition comprises:

from 0.1 to 99% by weight of component (a),  
from 0 to 25% by weight of component (b), and  
from 0 to 70% by weight of component (c).
14. The method according to claim 12, wherein said antimicrobial composition contains further GRAS flavoring agents selected from (d) phenols or their derivatives, (e) lipophilic esters, (f) terpenes, (g) acetals, (h) lipophilic aldehydes and (i) essential oils.

15. The method according to claim 12, wherein component (A) of said antimicrobial composition is benzyl alcohol.
16. The method according to claim 13, wherein said polyphenol compound is selected from the group consisting of:  
catechol, resorcinol, hydroquinone, phloroglucinol, pyrogallol, cyclohexane, resveratrol, usnic acid, acylpolyphenols, lignins, anthocyanins, flavones, catechols, gallic acid derivatives, caffeic acid, flavonoids, derivatives of the mentioned polyphenols, and extracts from Camellia, Primula; and said lipophilic GRAS acid is selected from the group consisting of:  
adipic acid, capronic acid, pelargonic acid, phenoxyacetic acid, valeric acid, iso-valeric acid, cinnamic acid, mandelic acid and their derivatives.
17. The method according to claim 12, wherein the component A of said antimicrobial composition comprises:  
  
from 0.1 to 99% by weight of benzyl alcohol;  
from 0 to 99.8% by weight of component (a<sub>i</sub>);  
from 0 to 25% by weight of component (b); and  
from 0 to 70% by weight of component (c).



18. The method according to claim 17, wherein said antimicrobial composition comprises from 0.001 to 25% by weight of further lipophilic GRAS flavoring agents selected from the group consisting of phenols or their derivatives, esters, terpenes, acetals, aldehydes and essential oils or extracts thereof.
19. The method according to claim 18, wherein said further lipophilic GRAS flavoring agents are phenols and/or essential oils or extracts thereof having a high content of alcohols, aldehydes, phenols, acetates or esters.
20. The method according to claim 1, wherein said antimicrobial composition does not contain any derivatives of the GRAS flavoring agents.
21. The method according to claim 17, wherein said antimicrobial composition comprises one or two lipophilic GRAS flavor alcohols and at least one polyphenol compound.
22. The method according to claim 21, wherein said polyphenol compound is tannin.

23. The method according to claim 22, wherein said antimicrobial composition contains from 20 to 98% by weight of benzyl alcohol and from 0.01 to 10% by weight of tannin.
24. The method according to claim 1, wherein said antimicrobial composition further comprises monohydric or polyhydric alcohols having from 2 to 10 carbon atoms, and additives selected from the group consisting of emulsifiers, stabilizers, antioxidants, preservatives, solvents and carriers.
25. The method according to claim 1, wherein said antimicrobial composition consists of GRAS flavoring agents.
26. The method according to claim 1, wherein said step of applying said antimicrobial composition to the surface of the plants is carried out by spraying, immersion or nebulizing.
27. A method for protecting plants from microbial attack shortly before or after the harvest, comprising the distribution of said antimicrobial composition within the plant by the step of adding the antimicrobial composition to nutrient media, nutrient liquids and/or water, said antimicrobial composition comprising at least two GRAS (generally recognized as safe) flavoring agents.

28. The method according to claim 27, wherein said GRAS flavoring agents are selected from the group consisting of (a) GRAS flavor alcohols or their derivatives, (b) polyphenol compounds, (c) GRAS flavor acids or their derivatives, (d) phenols or their derivatives, (e) esters, (f) terpenes, (g) acetals, (h) aldehydes and (i) essential oils.
29. The method according to claim 27, wherein said antimicrobial composition comprises at least one GRAS flavor alcohol.
30. The method according to claim 29, wherein said antimicrobial composition comprises less than 50% by weight of an alcohol selected from the group consisting of ethanol, isopropanol or benzyl alcohol or a mixture of these alcohols.
31. The method according to claim 27, wherein said antimicrobial composition comprises at least one hydrophilic alcoholic GRAS flavoring agent and/or one hydrophilic non-alcoholic GRAS flavoring agent.
32. The method according to claim 31, wherein said antimicrobial composition further comprises benzyl alcohol and/or a polyphenol compound.

33. The method according to claim 27, wherein said antimicrobial composition comprises:
- (A) at least one GRAS flavor alcohol (a) and/or its derivative; and
  - (B) at least one flavoring agent selected from the group consisting of (b) polyphenol compounds and (c) GRAS flavor acids or their derivatives.
34. The method according to claim 33, wherein said antimicrobial composition comprises:
- from 0.1 to 99% by weight of GRAS flavor alcohol;
  - from 0 to 25% by weight of polyphenol compounds; and
  - from 0 to 70% by weight of GRAS flavor acids or their derivatives.
35. The method according to claim 27, wherein said antimicrobial composition comprises benzyl alcohol and at least one further GRAS flavoring agent.
36. The method according to claim 35, wherein said further GRAS flavoring agents are selected from (a) GRAS flavor alcohols or their derivatives, (b) polyphenol compounds, (c) GRAS flavor acids or their derivatives, (d) phenols or their derivatives, (e) esters, (f) terpenes, (g) acetals, (h) aldehydes and (i) essential oils.

37. The method according to claim 36, wherein said antimicrobial composition comprises less than 50% by weight of benzyl alcohol or of a mixture of benzyl alcohol with ethanol and/or isopropanol.

38. The method according to claim 33, wherein said antimicrobial composition comprises:

benzyl alcohol as a necessary component;

at least one other GRAS flavor alcohol and/ or their derivatives;

at least one polyphenol compound; and/or

at least one GRAS acid and/or their derivatives.

39. The method according to claim 38, wherein said further GRAS flavor alcohol is selected from the group consisting of:

acetoin, ethyl alcohol, propyl alcohol, isopropyl alcohol, propylene

glycol, glycerol, n-butyl alcohol, iso-butyl alcohol, hexyl alcohol, L-

menthol, octyl alcohol, cinnamyl alcohol,  $\alpha$ -methylbenzyl alcohol,

heptyl alcohol, n-amyl alcohol, iso-amyl alcohol, anisalcohol, citronellol,

n-decyl alcohol, geraniol,  $\beta$ , $\gamma$ -hexenol, lauryl alcohol, linalool, nerolidol,

nonadienol, nonyl alcohol, rhodinol, terpineol, borneol, clineol, anisole,

cuminy alcohol, 10-un-decene-1-ol, 1-hexadecanol or their derivatives;

said polyphenol compound is selected from the group consisting of:

catechol, resorcinol, hydroquinone, phloroglucinol, pyrogallol, cyclohexane, resveratrol, usnic acid, acylpolyphenols, lignins, anthocyanins, flavones, catechols, gallic acid derivatives, caffeic acid, flavonoids, derivatives of the mentioned polyphenols, and extracts from Camellia, Primula; and

said GRAS acid is selected from the group consisting of:

acetic acid, aconitic acid, adipic acid, formic acid, malic acid, capronic acid, hydrocinnamic acid, pelargonic acid, lactic acid, phenoxyacetic acid, phenylacetic acid, valeric acid, iso-valeric acid, cinnamic acid, citric acid, mandelic acid, tartaric acid, fumaric acid, tannic acid and their derivatives.

40. The method according to claim 38, wherein said antimicrobial composition comprises:

from 0.1 to 99% by weight of benzyl alcohol;

from 0 to 99.8% by weight of other GRAS flavor alcohols and/or their derivatives;

from 0 to 25% by weight of polyphenol compounds; and

from 0 to 70% by weight of GRAS acids and/or their derivatives.

41. The method according to claim 38, wherein said antimicrobial composition comprises further GRAS flavoring agents selected from

the group consisting of phenols, esters, terpenes, acetals, aldehydes and essential oils.

42. The method according to claim 41, wherein said antimicrobial composition contains from 0.001 to 25% by weight of said further GRAS flavoring agents.
43. The method according to claim 42, wherein said further GRAS flavoring agents are phenols and/or essential oils.
44. The method according to claim 27, wherein said antimicrobial composition does not contain any derivatives of the GRAS flavoring agents.
45. The method according to claim 38, wherein said antimicrobial composition contains one or two GRAS flavor alcohols and at least one polyphenol compound.
46. The method according to claim 45, wherein said polyphenol compound is tannin.
47. The method according to claim 46, wherein said antimicrobial composition contains from 20 to 98% by weight of benzyl alcohol and from 0.01 to 10% by weight of tannin.

48. The method according to claim 27, wherein said antimicrobial composition is the composition of claim 12.
49. A method for protecting plants shortly before or after the harvest from insects and insect larvae, comprising the steps of:
- (i) applying an insecticidal composition to the surface of the plants and/or
  - (ii) distributing an insecticidal composition within the plant by adding the insecticidal composition to nutrient media, nutrient liquids and/or water; wherein said insecticidal composition is a composition containing GRAS flavoring agents as defined in claim 1.
50. The method according to claim 49, wherein the step of applying the insecticidal composition to the surface of the plant is carried out by spraying, immersion or nebulizing.
51. The method according to claim 1, wherein said plants are selected from the group consisting of cotton, cereals, rice, corn, potatoes, tobacco, coffee, cocoa, tea, vegetables, fruits, nuts, spices, herbs, seeds, ornamental plants, cultured flowers and flowers for cutting.



Page 14 of 14

PATENT  
Attorney Docket No. von Kreisler.021

### PLANT PROTECTION

The present invention relates to methods for protecting plants and/or plant parts from insects and insect larvae and from microbial attack, comprising the distribution or application of specific insecticidal or antimicrobial compositions to the surface of the plants and/or plant parts, and/or the distribution of the compositions within the plant and/or plant part; insecticides and biocides comprising said insecticidal or antimicrobial compositions; and the use of said insecticidal or antimicrobial compositions for the treatment of plants and/or plant parts.

During growth and especially after the harvest, plants are sensitive towards molds, bacteria, insects and insect larvae which adversely affect the quality of the plants or of the harvested plant materials or destroy them or make them useless. In addition, especially in plants used as food or feed products, there is a great problem in that molds and bacteria bring toxins onto the plants and harvested plant materials whose ingestion may be noxious to humans and animals. Insects and insect larvae can contaminate or destroy the plants especially following the harvest, namely during storage or in the course of processing. These problems are aggravated by the fact that the use of biocides and insecticides must be discontinued one week before the harvest on principle, so that an effective

antimicrobial and insecticidal protection generally is no longer available at the time of harvesting.

5 It is known that tea-tree oil and oregano extracts when applied to plants act as repellants, *i.e.*, exhibit some insecticidal property. In addition, from JP-A-62126931, it is known that vegetables can be sprayed for sterilization with an aqueous solution containing a flavonoid, an alcohol, such as ethanol, propylene glycol or glycerol, an organic acid and/or a calcium salt. However, these agents did not provide sufficient protection for the plants and plant parts from microbial and insect attacks. Further, WO 96/29895 and WO 98/58540 describe processing aids and additives for foodstuffs in which specific antimicrobial compositions containing at least two GRAS (generally recognized as safe) flavoring agents as antimicrobial components are employed.

15 It has been the object of the present invention to provide a method for providing sufficient antimicrobial and/or Insecticidal protection for plants, especially shortly before or after the harvest. Surprisingly, it has now been found that the compositions known from WO 96/29895 and WO 98/58540 have suitable antimicrobial properties which make them appear suitable for the present purpose. Especially, it was found that these compositions do not only act as repellants, but also hinder the growth or proliferation of insects and insect larvae *in vivo*. Finally, it was found among these compositions that particularly high antimicrobial and insecticidal activities are observed in those containing an

aromatic GRAS flavor alcohol, especially the compositions containing benzyl alcohol, and those containing both a lipophilic and a hydrophilic GRAS flavoring agent. Due to their being toxicologically safe, such compositions may also be used shortly before or after the harvest.

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Thus, the present application relates to:

(1) a method for protecting plants and/or plant parts from microbial attack, comprising the distribution or application of an antimicrobial composition to the surface of the plants and/or plant parts, said antimicrobial composition containing

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(i) at least one lipophilic GRAS (generally recognized as safe) flavoring agent; and

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(ii) at least one hydrophilic GRAS flavoring agent;

(2) a preferred embodiment of the method as defined in (1) wherein said antimicrobial composition exclusively consists of GRAS flavoring agents;

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(3) a method for protecting plants and/or plant parts from microbial attack, comprising the distribution of said antimicrobial composition within the plant and/or plant part, said antimicrobial composition within the plant and/or plant part, said

antimicrobial composition containing at least two GRAS (generally recognized as safe) flavoring agents;

(4) a preferred embodiment of the method as defined in (3) wherein said antimicrobial composition contains

(A) one or more GRAS flavor alcohols (a) or their derivatives; and

(B) one or more flavoring agents selected from

(b) polyphenol compounds; and

(c) GRAS flavor acids or their derivatives;

(5) a preferred embodiment of the methods as defined in (3) and (4) wherein said antimicrobial composition contains

(a1) benzyl alcohol as a necessary component; and optionally

(a2) one or more further GRAS flavor alcohols or their derivatives; and

(b) one or more polyphenol compounds; and/or

(c) one or more GRAS acids or their derivatives;

(6) a preferred embodiment of the method as defined in (3) to (5) wherein said antimicrobial composition is as defined in (1) and (2);

(7) a method for protecting plants and/or plant parts from insects and insect larvae, comprising:

(i) the distribution or application of an insecticidal composition to the surface of the plants and/or plant parts; and/or

(ii) the distribution of an insecticidal composition within the plant and/or plant part;

wherein said insecticidal composition is a composition containing GRAS flavoring agents as defined in (1) to (6);

(8) an insecticide and/or repellant, comprising an insecticidal composition as defined in (1) to (5);

(9) a biocide, especially fungicide and bactericide, comprising a composition as defined in (1) to (5), preferably as defined in (1) and (2);

(10) the use of the composition as defined in (1) to (5), preferably as defined in (1) and (2), for treating plants and/or plant parts.

The term "plants and/or plant parts" within the meaning of the present invention is to be understood as follows: During the nursing and growth to the harvest, the

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In detail, the following GRAS flavor alcohols may be employed, for example: benzyl alcohol, acetoin (acetylmethylcarbinol), ethyl alcohol (ethanol), propyl alcohol (1-propanol), isopropyl alcohol (2-propanol, isopropanol), propylene glycol, glycerol, n-butyl alcohol (n-propyl carbinol), iso-butyl alcohol (2-methyl-1-propanol), hexyl alcohol (hexanol), L-menthol, octyl alcohol (n-octanol), cinnamyl alcohol (3-phenyl-2-propene-1-ol),  $\alpha$ -methylbenzyl alcohol (1-phenyl-ethanol), heptyl alcohol (heptanol), n-amyl alcohol (1-pentanol), iso-amyl alcohol (3-methyl-1-butanol), anisalcohol (4-methoxybenzyl alcohol, p-anisalcohol), citronellol, n-decyl alcohol (n-decanol), geraniol,  $\beta,\gamma$ -hexenol (3-hexenol), lauryl alcohol (dodecanol), linalool, nerolidol, nonadienol (2,6-nonadiene-1-ol), nonyl alcohol (nonanol-1), rhodinol, terpineol, borneol, clineol (eucalyptol), anisole, cuminyl alcohol (cuminol), 10-undecene-1-ol, 1-hexadecanol. As said derivatives, both natural and synthetic (naturally occurring or not) derivatives can be employed. Suitable derivatives include, for example, the esters, ethers and carbonates of the above mentioned GRAS flavor alcohols. Particularly preferred GRAS flavor alcohols are benzyl alcohol, 1-propanol, glycerol, propylene glycol, n-butyl alcohol, citronellol, hexanol, linalool, acetoin and their derivatives.

As component (b), the following polyphenols may be employed:

catechol, resorcinol, hydroquinone, phloroglucinol, pyrogallol, cyclohexane, resveratrol, usnic acid, acylpolyphenols, lignins, anthocyanes, flavones, catechols, gallic acid derivatives (e.g., tannins, gallotannin, tannic acids, gallotannic acids), cranosol, carnosolic acid (including their derivatives, such as (2,5-dihydroxy-



phenyl)carboxylic and (2,5-dihydroxyphenyl)alkylenecarboxylic substitutions, salts, esters, amides); caffeic acid and its esters and amides, flavonoids (e.g., flavone, flavonol, isoflavone, gossypetin, myricetin, robinetin, apigenin, morin, taxifolin, eriodictyol, naringen, rutin, hesperidin, troxerutin, chrysin, tangeritin, luteolin, catechols, quercetin, fisetin, kaempferol, galangin, rotenoids, auronones, flavonols, diols), extracts, e.g., from Camellia, Primula. Further, their possible derivatives, e.g., salts, acids, esters, oxides and ethers, may also be used. A particularly preferred polyphenol is tannin (a GRAS compound).

As component (c), the following GRAS acids may be used, for example:

acetic acid, aconitic acid, adipic acid, formic acid, malic acid (1-hydroxysuccinic acid), capronic acid, hydrocinnamic acid (3-phenyl-1-propionic acid), pelargonic acid (nonanoic acid), lactic acid (2-hydroxypropionic acid), phenoxyacetic acid (glycolic acid phenyl ether), phenylacetic acid ( $\alpha$ -toluenic acid), valeric acid (pentanoic acid), iso-valeric acid (3-methylbutyric acid), cinnamic acid (3-phenylpropenoic acid), citric acid, mandelic acid (hydroxyphenylacetic acid), tartaric acid (2,3-dihydroxybutanedioic acid; 2,3-dihydroxysuccinic acid), fumaric acid, tannic acid and their derivatives.

Suitable derivatives of the GRAS flavor acids according to the present invention are esters (e.g., C<sub>1-6</sub> alkyl esters and benzyl esters), amides (including N-substituted amides) and salts (alkali, alkaline earth and ammonium salts) of the above mentioned acids. According to the present invention, the term "derivatives"

also encompasses modifications of the side-chain hydroxy functions (e.g., acyl and alkyl derivatives) and modifications of the double bonds (e.g., the perhydrogenated and hydroxylated derivatives of the mentioned acids).

5 As component (d), the following phenol compounds may be employed:

thymol, methyleugenol, acetyeugenol, safroi, eugenol, isoeugenol, anethole, phenol, methylchavicol (estragol; 3-(4-methoxyphenyl)-1-propene), carvacrol,  $\alpha$ -bisabolol, fornesol, anisole (methoxybenzene), propenylguaethol (5-propenyl-2-ethoxyphenol) and their derivatives. Derivatives within the meaning of the present  
10 invention are compounds in which the phenolic hydroxy group is esterified or etherified.

As GRAS esters (component (e)), for example, allicin and the following acetates may be used: iso-amyl acetate (3-methyl-1-butyl acetate), benzyl acetate, benzylphenyl acetate, n-butyl acetate, cinnamyl acetate (3-phenylpropenyl acetate), citronellyl acetate, ethyl acetate (acetic ester), eugenol acetate (acetyeugenol), geranyl acetate, hexyl acetate (hexanyl ethanoate), hydrocinnamyl acetate (3-phenylpropyl acetate), linalyl acetate, octyl acetate, phenylethyl acetate, terpinyl acetate, triacetin (glyceryl triacetate), potassium  
15 acetate, sodium acetate and calcium acetate. Further suitable esters are the ester derivatives of the above defined acids (component (b2)).  
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As terpenes (component (f)), there may be used, for example, camphor, limonene and  $\beta$ -caryophyllene.

The acetals (component (g)) which can be used include, e.g., acetal, acetaldehyde dibutyl acetal, acetaldehyde dipropyl acetal, acetaldehyde phenethyl propyl acetal, cinnamic aldehyde ethylene glycol acetal, decanal dimethyl acetal, heptanal dimethyl acetal, heptanal glyceryl acetal and benzaldehyde propylene glycol acetal.

As aldehydes (component (h)), there may be used, e.g., acetaldehyde, anisaldehyde, benzaldehyde, iso-butyl aldehyde (methyl-1-propanal), citral, citronellal, n-caprylic aldehyde (n-decanal), ethylvanillin, furfural, heliotropin (piperonal), heptyl aldehyde (heptanal), hexyl aldehyde (hexanal), 2-hexenal ( $\beta$ -propyl-acrolein), hydrocinnamic aldehyde (3-phenyl-1-propanal), lauryl aldehyde (do-decanal), nonyl aldehyde (n-nonanal), octyl aldehyde (n-octanal), phenylacetaldehyde (1-oxo-2-phenylethane), propionaldehyde (propanal), vanillin, cinnamic aldehyde (3-phenylpropenal), perillaldehyde and cuminaldehyde.

The following essential oils and/or alcoholic or glycolic extracts or extracts obtained by CO<sub>2</sub> high-pressure processes from the mentioned plants (component (i)) can also be employed according to the invention:

(i1) oils or extracts having a high content of alcohols: melissa, coriander, cardamon, eucalyptus;

(i2) oils or extracts having a high content of aldehydes: Eucalyptus citriodora, cinnamon, lemon, lemon grass, melissa, citronella, lime, orange;

5 (i3) oils or extracts having a high content of phenols: origanum, thyme, rosemary, orange, clove, fennel, camphor, mandarin, anise, cascarrilla, estragon and pimento;

(i4) oils or extracts having a high content of acetates: lavender;

(i5) oils or extracts having a high content of esters: mustard, onion, garlic;

10 (i6) oils or extracts having a high content of terpenes: pepper, bitter orange, caraway, dill, lemon, peppermint, nutmeg.

In the following, the antimicrobial composition used in the above defined method (1) is further illustrated. In this composition, the lipophilic GRAS flavoring agents are preferably selected from (a<sub>1</sub>) lipophilic GRAS flavor alcohols or their derivatives, (b) polyphenol compounds, (c<sub>1</sub>) lipophilic GRAS flavor acids or their derivatives, (d) phenols or their derivatives, (e<sub>1</sub>) lipophilic esters, (f) terpenes, (g) acetals, (h<sub>1</sub>) lipophilic aldehydes and (i) essential oils. The antimicrobial composition preferably contains two of the mentioned GRAS flavoring agents.

20 Suitable lipophilic GRAS flavor alcohols (a<sub>1</sub>) among the defined alcohols (a) include, in particular:

aromatic GRAS flavor alcohols, including benzyl alcohol, 2-phenylethanol, 1-phenylethanol, cinnamyl alcohol, hydrocinnamyl alcohol, 1-phenyl-1-propanol and anisalcohol, and aliphatic GRAS flavor alcohols, including n-butyl alcohol, iso-butyl alcohol, hexyl alcohol, L-menthol, octyl alcohol, heptyl alcohol, n-amyl alcohol, iso-amyl alcohol, anisalcohol, citronellol, n-decyl alcohol, geraniol,  $\beta$ , $\gamma$ -hexenol, lauryl alcohol, linalool, nerolidol, nonadienol, nonyl alcohol, rhodinol, terpeneol, borneol, clineol, anisole, cuminyl alcohol, 10-undecene-1-ol and 1-hexadecanol and their derivatives. The aromatic GRAS flavor alcohols, especially benzyl alcohol, are preferred.

According to the present invention, the hydrophilic GRAS flavoring agent is a hydrophilic alcoholic GRAS flavoring agent ( $a_h$ ) or a hydrophilic non-alcoholic GRAS flavoring agent, wherein said hydrophilic alcoholic GRAS flavoring agent ( $a_h$ ) is preferably a monohydric or polyhydric alcohol having from 2 to 10, more preferably from 2 to 7, carbon atoms, especially one selected from acetoin, ethyl alcohol, propyl alcohol, isopropyl alcohol, propylene glycol and glycerol, and said hydrophilic non-alcoholic GRAS flavoring agent is a hydrophilic organic GRAS flavor acid ( $c_h$ ) having from 1 to 15 carbon atoms or a physiological salt thereof, a hydrophilic acetate ( $e_h$ ) or a hydrophilic aldehyde ( $h_h$ ). Preferred hydrophilic organic acids ( $c_h$ ) include those which contain from 2 to 10 carbon atoms, especially acetic acid, aconitic acid, formic acid, malic acid, lactic acid, phenylacetic acid, citric acid, mandelic acid, tartaric acid, fumaric acid, tannic acid, hydrocinnamic acid and their physiological salts. Said hydrophilic acetate ( $e_h$ ) is

preferably allicin, triacetin, potassium acetate, sodium acetate and calcium acetate. Said hydrophilic aldehyde ( $h_h$ ) is preferably selected from furfural, propionaldehyde and vanillin.

The lipophilic polyol compound (b), phenols or their derivatives (d), terpenes (f), acetals (g) and essential oils (i) in the composition of method (1) are preferably the above defined compounds (b), (d), (f), (g) and (i). The lipophilic GRAS flavor acids or their derivatives ( $c_l$ ), lipophilic esters ( $e_l$ ) and lipophilic aldehydes include all specifically mentioned acids, esters and aldehydes, except for the compounds ( $c_h$ ), ( $e_h$ ) and ( $h_h$ ) specifically mentioned above.

In a preferred embodiment of method (1), the antimicrobial composition contains either:

(i) two lipophilic GRAS flavor alcohols ( $a_l$ ), but no benzyl alcohol and no polyphenol compounds (b); or

(ii) benzyl alcohol and/or a polyphenol compound (b), but no further GRAS flavor alcohols.

It is particularly preferred for the antimicrobial composition to contain exclusively non-alcoholic hydrophilic GRAS flavoring agents, especially exclusively a hydrophilic GRAS flavor acid ( $C_h$ ), and for the antimicrobial composition to contain

from 0.01 to 99% by weight, preferably from 0.1 to 90% by weight, of benzyl alcohol or polyphenol compounds (b) and from 0.01 to 50% by weight, preferably from 0.1 to 30% by weight, of hydrophilic non-alcoholic GRAS flavoring agents.

In a further preferred embodiment of method (1), the antimicrobial composition contains:

(A) one or more GRAS flavor alcohols (a) or their derivatives; and

(B) one or more flavoring agents selected from polyphenol compounds (b) and lipophilic GRAS flavor acids or their derivatives (c).

It is preferred for the composition to contain from 0.1 to 99% by weight, preferably from 0.5 to 99% by weight, of component (a), from 0 to 25% by weight, preferably from 0.01 to 10% by weight, of component (b), and from 0 to 70% by weight, preferably from 0.01 to 30% by weight, of component (c).

In addition, the antimicrobial composition may contain further GRAS flavoring agents selected from (d) phenols or their derivatives, (e<sub>1</sub>) lipophilic esters, (f) terpenes, (g) acetals, (h<sub>1</sub>) lipophilic aldehydes and (i) essential oils.

It is further preferred for component (A) of the antimicrobial composition to contain benzyl alcohol as a necessary component (a<sub>1</sub>) and optionally one or more further

lipophilic GRAS flavor alcohols or their derivatives (a<sub>1</sub>). Preferably, this antimicrobial composition contains:

from 0.1 to 99% by weight, preferably from 0.1 to 75% by weight, of benzyl alcohol;

from 0 to 99.8% by weight, preferably from 0.01 to 99% by weight, of component (a<sub>1</sub>); and

from 0 to 25% by weight, preferably from 0.01 to 10% by weight, of component (b);

from 0 to 70% by weight, preferably from 0.01 to 30% by weight, of component (c).

The composition employed may contain further lipophilic GRAS flavoring agents (d) to (i) as defined above, preferably from 0.001 to 25% by weight, more preferably from 0.01 to 9% by weight, of said further GRAS flavoring agents (d) to (i). Said further lipophilic GRAS flavoring agents more preferably include phenols (d) and/or essential oils (i).

In a further particularly preferred embodiment of method (1), component (A) of the antimicrobial composition consists of two lipophilic GRAS flavor alcohols, and component (B) contains at least one polyphenol compound (b). Said polyphenol compound (b) is preferably tannin, particularly preferred being a composition



which contains from 20 to 98% by weight of benzyl alcohol and from 0.01 to 10% by weight of tannin.

In the following, preferred embodiments of the antimicrobial/insecticidal composition employed in methods (3) and (7) are illustrated in more detail:  
5 Preferably, the antimicrobial/insecticidal composition contains at least one GRAS flavor alcohol (a), especially benzyl alcohol. Preferred are those compositions which contain less than 50% by weight, preferably less than 30% by weight, more preferably less than 20% by weight, of ethanol, isopropanol or benzyl alcohol or  
10 a mixture of these substances.

In another preferred embodiment of methods (3) and (7), the antimicrobial/insecticidal composition contains at least one hydrophilic alcoholic GRAS flavoring agent and/or one hydrophilic non-alcoholic GRAS flavoring agent.  
15 The proportion of hydrophilic alcoholic GRAS flavoring agents may be up to 99% by weight of the insecticidal composition and is preferably from 30 to 98% by weight, more preferably from 80 to 95% by weight. The proportion of hydrophilic non-alcoholic GRAS flavoring agents in the insecticidal composition may be up to 90% by weight and is preferably from 0.1 to 50% by weight. Preferred are  
20 those compositions which further contain benzyl alcohol and/or a polyphenol compound (b) in addition to the mentioned hydrophilic compounds.

Hydrophilic alcoholic GRAS flavoring agents are the above defined compounds ( $a_h$ ). Hydrophilic non-alcoholic GRAS flavoring agents include, in particular, the above defined organic acids ( $c_h$ ).

5 Further preferably employed antimicrobial/insecticidal compositions are the compositions stated above under (4) and (5). The composition as defined under (4) may contain:

from 0.1 to 99.9% by weight, preferably from 0.5 to 99% by weight, of component (a);

10 from 0 to 25% by weight, preferably from 0.01 to 10% by weight, of component (b); and

from 0 to 70% by weight, preferably from 0.01 to 30% by weight, of component (c).

15 In this embodiment of the invention, component (a) contains one or more GRAS flavor alcohols. Preferred is the use of two or three GRAS flavor alcohols. The mixing ratio of component (a) to component (b) is preferably between 10,000 : 1 and 1 : 10,000, more preferably between 1000 : 1 and 1 : 1000, and still more preferably between 100 : 1 and 1 : 100.

20 In the method according to the invention as defined above under (4), the composition may contain further GRAS flavoring agents, such as the above defined GRAS flavor alcohols or their derivatives (a) with the exception of benzyl

alcohol, polyphenol compounds (b), acids (c), phenols (d), esters (e), terpenes (f), acetals (g), aldehydes (h) and essential oils (i).

Preferred antimicrobial/insecticidal compositions according to the present invention are those which contain less than 50% by weight, preferably less than 30% by weight, more preferably less than 20% by weight, of benzyl alcohol or of a mixture of benzyl alcohol with ethanol and/or isopropanol.

In a further preferred embodiment of the present invention, the antimicrobial/insecticidal composition contains at least one hydrophilic alcoholic GRAS flavoring agent and/or one hydrophilic non-alcoholic GRAS flavoring agent. As to the proportion of the hydrophilic compound and as to particularly preferred hydrophilic compounds, reference is made to the above indications. Particularly preferred in this connection are those antimicrobial/insecticidal compositions which contain polyphenol compounds in addition to benzyl alcohol and the mentioned hydrophilic GRAS flavoring agents.

As mentioned above under (5), in a particularly preferred embodiment of the method according to the invention, the insecticidal or antimicrobial composition contains:

(a1) benzyl alcohol as a necessary component; and optionally

(a2) one or more further GRAS flavor alcohols or their derivatives; and

- (b) one or more polyphenol compounds; and/or
- (c) one or more GRAS acids or their derivatives.

Suitable amounts of components (a1), (a2), (b) and (c) in the above defined  
5 insecticidal and antimicrobial composition are:

from 0.1 to 99% by weight, preferably from 0.1 to 75% by weight, of benzyl  
alcohol;

10 from 0 to 99.8% by weight, preferably from 0.01 to 99% by weight, of component  
(a2);

from 0 to 25% by weight, preferably from 0.01 to 10% by weight, of component  
(b1); and/or

from 0 to 70% by weight, preferably from 0.01 to 30% by weight, of component  
(b2).

15 The particularly preferred insecticidal and antimicrobial composition may further  
contain the above mentioned components (d) to (i), which are also GRAS  
flavoring agents.

20 The proportion of components (d) to (i) in the antimicrobial composition is  
preferably equal to or smaller than 25% by weight, preferably within a range of  
from 0.001 to 9% by weight. Preferred among the further GRAS flavoring agents  
are the phenols (d) and the essential oils (i).

Most preferred for methods (3) and (7) of the present invention are those antimicrobial or insecticidal compositions which correspond to the compositions defined above under (1) and (2), *i.e.*, compositions containing at least one hydrophilic GRAS flavoring agent, wherein the component thereof having insecticidal or antimicrobial activity optionally consists exclusively of GRAS flavoring agents, *i.e.*, does not contain any derivatives of the GRAS flavoring agents. As an example of such a composition, there may be mentioned a mixture of benzyl alcohol, one or two of the above mentioned GRAS flavor alcohols (a2) and tannic acid. Such mixture preferably contains from 20 to 98% by weight of benzyl alcohol and from 0.01 to 10% by weight of tannic acid. Another example of a preferred composition is a mixture of 2 alcohols, a polyphenol (especially tannic acid) and an essential oil (especially a phenolic essential oil, component (h3)).

In addition to components (a) to (i), the antimicrobial/insecticidal compositions may additionally contain further compounds (j), such as alcohols (j1), emulsifiers (j2), stabilizers (j3), antioxidants (j4), preservatives (j5), solvents (j6), carriers (j7) *etc.*

The solvents (j6) include, for example, vegetable materials including rapeseed oil, soybean oil, edible fatty acids *etc.*, and chemical solvents including nitroturpentine, polyurethanes, aliphatic hydrocarbons, isoparaffin *etc.*, and mixtures thereof.

5 According to the invention, the alcohols (j1) are monohydric or polyhydric alcohols having from 2 to 10 carbon atoms, preferably from 2 to 7 carbon atoms, not including the GRAS alcohols (a). Preferably, such amounts of GRAS flavor alcohols (a) and further alcohols (j1) are employed that their mixing ratio is between 1000 : 1 and 1 : 1000, especially between 100 : 1 and 1 : 100, more  
10 preferably between 10 : 1 and 1 : 10.

It is particularly preferred in the method according to the present invention to use systems which exclusively consist of GRAS flavoring agents, especially when the treated plants and/or plant parts are later ingested as foods, beverages or luxuries or otherwise come into contact with the human body, because this prevents contamination of the plants and/or plant parts with non-GRAS compounds. Further, it should be taken care that the insecticidal and antimicrobial composition is free of ethanol and isopropanol, or free of noxious doses of ethanol and isopropanol, since these substances can be absorbed by the plants and/or plant parts, and can be inhaled by the persons who further process such plants. In addition, there may be a danger of explosion when these compounds are used.

The method according to the invention is suitable for the treatment of plants during nursing and growth, especially shortly before the harvest (*e.g.*, during the last week before the harvest) and also after the harvesting of the plants. The insecticidal and/or antimicrobial treatment may be both by application to the surface of the plants and/or plant parts (*e.g.*, by spraying, immersion, nebulizing, *etc.*) and during growth of the plants by adding the insecticidal or antimicrobial composition to nutrient media, nutrient liquids, water *etc.* In the latter way, the insecticidal or antimicrobial composition is distributed within the plant or plant part. The insecticidal and/or antimicrobial composition can be contacted with the plants both in undiluted form and diluted in aqueous solvent systems, organic solvent systems and/or oil suspensions.

It could be shown that concentrations of the insecticidal and/or antimicrobial composition within a range of from 0.001 to 100 mg/g of plant, preferably from 0.1 to 10 mg/g of plant, ensure sufficient insecticidal and antimicrobial protection when the composition is applied to plant surfaces. When applied in nutrient media and nutrient solutions, a concentration of the antimicrobial composition of from 0.001 to 100, preferably from 0.1 to 10 mg, per g of nutrient medium or nutrient solution is sufficient. For economical reasons, it is a matter of course that as low as possible a concentration of the insecticidal or antimicrobial composition will be employed.

In particular, the method according to the invention is suitable for the treatment of, for example, cotton, cereals, rice, corn, potatoes, tobacco, coffee, tea, vegetables, fruits, seeds of the mentioned plants, nuts, spices, herbs, ornamental plants, cultured flowers and flowers for cutting, and for plant cultivation.



Especially, the method according to the invention significantly reduces or even completely suppresses the microorganisms, agents of decay, mycotoxin formers and parasites stated in the following list.

<b><u>Molds</u></b>  mildew species rust fungi leaf spot fungi <i>Fusarium</i> species <i>Aspergillus</i> species <i>Penicillium</i> species  <i>Rhizoctonia</i> <i>Peronospora</i> <i>Phytophthora</i> <i>Botrytis cinerea</i> <i>Rhizoctonia solani</i>  <i>Aspergillus ocraceus</i> <i>Aspergillus niger</i> <i>Clavosporium fusarium</i> <i>Penicilliums</i>	<b><u>Culture/plant (illustrative)</u></b>  cereals (field fungi) cereals (field fungi) cereals (field fungi) cereals (field/storage fungi) cereals (storage fungi) cereals (storage fungi)  tobacco, rape tobacco tobacco tobacco rice  coffee coffee coffee coffee
<b><u>Parasites</u></b>  Lepidoptera Lepidoptera ( <i>Chilo suppressalis</i> ) ( <i>Chaphalocrosis medinalis</i> ) ( <i>Ostrina nubilalis</i> ) Myzus persicase ( <i>Chaphalocrosis medinalis</i> )	<b><u>Culture/plant (illustrative)</u></b>  tomatoes; cotton rice rice corn tobacco
<b><u>Viruses</u></b>  tomato mosaic virus X virus Y virus rice stripe virus TYM virus Rhizomania BNYVV	<b><u>Culture/plant (illustrative)</u></b>  tomatoes potatoes potatoes rice rape sugar beet sugar beet

In addition to the insecticidal or antimicrobial composition, the insecticides and biocides (*i.e.*, bactericides, fungicides, virucides, sporicides) according to the invention may contain further compounds, such as the above mentioned compounds (i1) to (i7). In addition, the insecticides and biocides according to the invention may also be in the form of retard preparations. Such retard preparations are preferred, in particular, for use in nutrient media in order to thereby ensure as long as possible an insecticidal or biocidal activity. Suitable retard preparations include, for example, microcapsules or coatings in which the active substance is encapsulated or coated with a suitable encapsulating or coating material, such as cellulose derivatives.

The present invention is further illustrated by means of the following Examples. Further useful insecticidal and antimicrobial compositions in addition to the composition of the following Example are mentioned in WO 96/29859 and WO 98/58540, which are included herein by reference.

## Examples

The following culture plants are sprayed with an insecticidal/antimicrobial composition which contains the following components (in % by weight):

- 5      10.0% polyphenol (here: tannin)
- 18.2% benzyl alcohol
- 60.0% propylene glycol
- 8.0% lactic acid
- 3./8% essential oil (here: a phenol-containing essential oil)

10

The results are summarized in the following Tables 1 to 3.

**Table 1: Wheat**

	50% solution in	Spraying agent	Molds Section 35 LMBG (German Food and Consumer Goods Act) (S.A.)	Parasite larvae (S.A.)
W-0		blank, untreated	$5 \times 10^4/\text{g}$	populated 6 months after harvest
W-1	water	0.001 mg/g	$7 \times 10^3/\text{g}$	no growth
W-2	water	0.01 mg/g	$1 \times 10^3/\text{g}$	no growth
W-3	water	0.1 mg/g	$6 \times 10^2/\text{g}$	no growth
W-4	water	1 mg/g	$7 \times 10^1/\text{g}$	no growth
W-5	water	10 mg/g	$2 \times 10^1/\text{g}$	no growth
W-6	water	100 mg/g	$< 10/\text{g}$	no growth
W-7	solvent*	0.001 mg/g	$5 \times 10^3/\text{g}$	no growth
W-8	solvent*	0.01 mg/g	$4 \times 10^3/\text{g}$	no growth
W-9	solvent*	0.1 mg/g	$3 \times 10^2/\text{g}$	no growth
W-10	solvent*	1 mg/g	$6 \times 10^1/\text{g}$	no growth
W-11	solvent*	10mg/g	$3 \times 10^1/\text{g}$	no growth
W-12	solvent*	100mg/g	$1 \times 10^1/\text{g}$	no growth

\* here: rapeseed oil

**Table 2: Tobacco**

	50% solution in	Spraying agent	Molds Section 35 LMBG (German Food and Consumer Goods Act) (S.A.)	Parasite larvae (S.A.)
T-0		blank, untreated	6 x 10 <sup>4</sup> /g	populated 6 months after harvest
T-1	water	0.001 mg/g	5 x 10 <sup>4</sup> /g	no growth
T-2	water	0.01 mg/g	1 x 10 <sup>4</sup> /g	no growth
T-3	water	0.1 mg/g	8 x 10 <sup>3</sup> /g	no growth
T-4	water	1 mg/g	4 x 10 <sup>2</sup> /g	no growth
T-5	water	10 mg/g	6 x 10 <sup>1</sup> /g	no growth
T-6	water	100 mg/g	< 10/g	no growth
T-7	solvent*	0.001 mg/g	6 x 10 <sup>4</sup> /g	no growth
T-8	solvent*	0.01 mg/g	8 x 10 <sup>3</sup> /g	no growth
T-9	solvent*	0.1 mg/g	2 x 10 <sup>3</sup> /g	no growth
T-10	solvent*	1 mg/g	4 x 10 <sup>2</sup> g	no growth
T-11	solvent*	10 mg/g	6 x 10 <sup>1</sup> /g	no growth
T-12	solvent*	100 mg/g	4 x 10 <sup>1</sup> /g	no growth

\* here: rapeseed oil

**Table 3: Raw coffee**

	50% solution in	Spraying agent	Molds Section 35 LMBG (German Food and Consumer Goods Act) (S.A.)	Parasite larvae
C-0		blank, untreated	$4 \times 10^4/\text{g}$	populated 6 months after harvest
C-1	water	0.001 mg/g	$9 \times 10^3/\text{g}$	no growth
C-2	water	0.01 mg/g	$4 \times 10^3/\text{g}$	no growth
C-3	water	0.1 mg/g	$7 \times 10^3/\text{g}$	no growth
C-4	water	1 mg/g	$1 \times 10^3/\text{g}$	no growth
C-5	water	10 mg/g	$3 \times 10^2/\text{g}$	no growth
C-6	water	100 mg/g	$8 \times 10^1/\text{g}$	no growth
C-7	solvent*	0.001 mg/g	$2 \times 10^4/\text{g}$	no growth
C-8	solvent*	0.01 mg/g	$3 \times 10^3/\text{g}$	no growth
C-9	solvent*	0.1 mg/g	$8 \times 10^2/\text{g}$	no growth
C-10	solvent*	1 mg/g	$4 \times 10^2/\text{g}$	no growth
C-11	solvent*	10 mg/g	$6 \times 10^1/\text{g}$	no growth
C-12	solvent*	100 mg/g	$< 10/\text{g}$	no growth

\* here: rapeseed oil

**CLAIMS:**

(amended September 4, 2001)

1. A method for protecting plants shortly before or after the harvest from microbial attack, comprising the distribution or application of an antimicrobial composition to the surface of the plants, said antimicrobial composition containing

(ii) at least one lipophilic GRAS (generally recognized as safe) flavoring agent; and

(ii) at least one hydrophilic GRAS flavoring agent.

2. The method according to claim 1, wherein said lipophilic GRAS flavoring agents are selected from (a<sub>1</sub>) lipophilic GRAS flavor alcohols or their derivatives, (b) polyphenol compounds, (c<sub>1</sub>) lipophilic GRAS flavor acids or their derivatives, (d) phenols or their derivatives, (e<sub>1</sub>) lipophilic esters, (f) terpenes, (g) acetals, (h<sub>1</sub>) lipophilic aldehydes and (i) essential oils.

3. The method according to claim 1 or 2, wherein said antimicrobial composition contains at least two lipophilic GRAS flavoring agents, preferably two lipophilic GRAS flavor alcohols (a<sub>1</sub>).

4. The method according to claim 2 or 3, wherein said lipophilic GRAS flavor alcohols are selected from: aromatic GRAS flavor alcohols, including

benzyl alcohol, 2-phenylethanol, 1-phenylethanol, cinnamyl alcohol, hydrocinnamyl alcohol, 1-phenyl-1-propanol and anisalcohol, and aliphatic GRAS flavor alcohols, including n-butyl alcohol, iso-butyl alcohol, hexyl alcohol, L-menthol, octyl alcohol, heptyl alcohol, n-amyl alcohol, iso-amyl alcohol, anisalcohol, citronellol, n-decyl alcohol, geraniol,  $\beta$ , $\gamma$ -hexenol, lauryl alcohol, linalool, nerolidol, nonadienol, nonyl alcohol, rhodinol, terpeneol, borneol, clineol, anisole, cuminyl alcohol, 10-undecene-1-ol and 1-hexadecanol and their derivatives, said aromatic GRAS flavor alcohols, especially benzyl alcohol, being preferred.

5. The method according to one or more of claims 1 to 4, wherein said hydrophilic GRAS flavoring agent is a hydrophilic alcoholic GRAS flavoring agent ( $a_h$ ) or a hydrophilic non-alcoholic GRAS flavoring agent, wherein said hydrophilic alcoholic GRAS flavoring agent ( $a_h$ ) is preferably a monohydric or polyhydric alcohol having from 2 to 10, more preferably from 2 to 7, carbon atoms, especially one selected from acetoin, ethyl alcohol, propyl alcohol, isopropyl alcohol, propylene glycol and glycerol, and said hydrophilic non-alcoholic GRAS flavoring agent is a hydrophilic organic GRAS flavor acid ( $C_h$ ) having from 1 to 15 carbon atoms or a physiological salt thereof, a hydrophilic acetate ( $e_h$ ) or a hydrophilic aldehyde ( $h_h$ ).

6. The method according to claim 5, wherein said hydrophilic organic acid ( $C_h$ ) has from 2 to 10 carbon atoms, especially being selected from acetic



acid, aconitic acid, formic acid, malic acid, lactic acid, phenylacetic acid, citric acid, mandelic acid, tartaric acid, fumaric acid, tannic acid, hydrocinnamic acid and their physiological salts; said hydrophilic acetate ( $e_h$ ) is selected from allicin, triacetin, potassium acetate, sodium acetate and calcium acetate; and/or said hydrophilic aldehyde ( $h_h$ ) is selected from furfural, propionaldehyde and vanillin.

7. The method according to claim 5, wherein said antimicrobial composition contains less than 50% by weight, preferably less than 30% by weight, more preferably less than 20% by weight, of benzyl alcohol or of a mixture of benzyl alcohol with ethanol and/or isopropanol.
8. The method according to claim 5 or 6, wherein said antimicrobial composition contains two lipophilic GRAS flavor alcohols ( $a_l$ ), but no benzyl alcohol and no polyphenol compounds (b).
9. The method according to claim 5 or 6, wherein said antimicrobial composition contains benzyl alcohol and/or a polyphenol compound (b), but no further GRAS flavor alcohols.
10. The method according to claim 8 or 9, wherein said antimicrobial composition exclusively contains non-alcoholic hydrophilic GRAS flavoring agents, especially exclusively a hydrophilic GRAS flavor acid ( $C_h$ ).

11. The method according to claim 9 or 10, wherein said antimicrobial composition contains from 0.01 to 99% by weight, preferably from 0.1 to 90% by weight, of benzyl alcohol or polyphenol compounds (b) and from 0.01 to 50% by weight, preferably from 0.1 to 30% by weight, of hydrophilic non-alcoholic GRAS flavoring agents.

12. The method according to claim 1 or 2, wherein said antimicrobial composition contains  
(A) one or more GRAS flavor alcohols (a) or their derivatives; and  
(B) one or more flavoring agents selected from polyphenol compounds (b) and lipophilic GRAS flavor acids or their derivatives (c).

13. The method according to claim 12, wherein said antimicrobial composition contains

from 0.1 to 99% by weight, preferably from 0.5 to 99% by weight, of component (a),  
from 0 to 25% by weight, preferably from 0.01 to 10% by weight, of component (b), and  
from 0 to 70% by weight, preferably from 0.01 to 30% by weight, of component (c).

14. The method according to claim 12 or 13, wherein said antimicrobial composition contains further GRAS flavoring agents selected from (d) phenols or their derivatives, (e<sub>1</sub>) lipophilic esters, (f) terpenes, (g) acetals, (h<sub>1</sub>) lipophilic aldehydes and (i) essential oils.

5

15. The method according to one or more of claims 12 to 14, wherein component (A) of said antimicrobial composition contains benzyl alcohol as a necessary component (a1) and optionally one or more further lipophilic GRAS flavor alcohols or their derivatives (a<sub>i</sub>).

10

16. The method according to one or more of claims 12 to 15, wherein said polyphenol compound (b) is selected from:

catechol, resorcinol, hydroquinone, phloroglucinol, pyrogallol, cyclohexane, resveratrol, usnic acid, acylpolyphenols, lignins, anthocyanins, flavones, catechols, gallic acid derivatives, caffeic acid, flavonoids, derivatives of the mentioned polyphenols, and extracts from Camellia, Primula; and said lipophilic GRAS acid (c) is selected from:

15

adipic acid, capronic acid, pelargonic acid, phenoxyacetic acid, valeric acid, iso-valeric acid, cinnamic acid, mandelic acid and their derivatives.

20

17. The method according to claim 15 or 16, wherein said antimicrobial composition contains

from 0.1 to 99% by weight, preferably from 0.1 to 75% by weight, of benzyl alcohol;

from 0 to 99.8% by weight, preferably from 0.01 to 99% by weight, of component (a<sub>1</sub>); and

from 0 to 25% by weight, preferably from 0.01 to 10% by weight, of component (b);

from 0 to 70% by weight, preferably from 0.01 to 30% by weight, of component (c).

18. The method according to claim 17, wherein said antimicrobial composition contains further lipophilic GRAS flavoring agents (d) to (i), preferably from 0.001 to 25% by weight, more preferably from 0.01 to 9% by weight, of said further GRAS flavoring agents (d) to (i).

19. The method according to claim 18, wherein said further lipophilic GRAS flavoring agents are phenols (d) and/or essential oils (i).

20. The method according to one or more of claims 1 to 19, wherein said antimicrobial composition does not contain any derivatives of the GRAS flavoring agents.

21. The method according to one or more of claims 15 to 20, wherein said antimicrobial composition contains one or two lipophilic GRAS flavor alcohols (a<sub>1</sub>) and at least one polyphenol compound (b).

5 22. The method according to claim 21, wherein said polyphenol compound (b) is tannin.

10 23. The method according to claim 22, wherein said antimicrobial composition contains from 20 to 98% by weight of benzyl alcohol and from 0.01 to 10% by weight of tannin.

15 24. The method according to one or more of claims 1 to 23, wherein said antimicrobial composition further contains monohydric or polyhydric alcohols having from 2 to 10 carbon atoms, emulsifiers, stabilizers, antioxidants, preservatives, solvents and/or carriers.

25. The method according to one or more of claims 1 to 23, wherein said antimicrobial composition exclusively consists of GRAS flavoring agents.

20 26. The method according to one or more of claims 1 to 25, wherein said distribution or application comprises application of said antimicrobial composition to the surface of the plants, especially by spraying, immersion or nebulizing.

27. A method for protecting plants from microbial attack shortly before or after the harvest, comprising the distribution of said antimicrobial composition within the plant, which is effected by adding the antimicrobial composition to nutrient media, nutrient liquids and/or water, said antimicrobial composition containing at least two GRAS (generally recognized as safe) flavoring agents.
28. The method according to claim 27, wherein said GRAS flavoring agents are selected from (a) GRAS flavor alcohols or their derivatives, (b) polyphenol compounds, (c) GRAS flavor acids or their derivatives, (d) phenols or their derivatives, (e) esters, (f) terpenes, (g) acetals, (h) aldehydes and (i) essential oils.
29. The method according to claim 27 or 28, wherein said antimicrobial composition contains at least one GRAS flavor alcohol (a), preferably an aromatic GRAS flavor alcohol, especially benzyl alcohol.
30. The method according to claim 29, wherein said antimicrobial composition contains less than 50% by weight, preferably less than 30% by weight, more preferably less than 20% by weight, of ethanol, isopropanol or benzyl alcohol or a mixture of these substances.

31. The method according to claim 27 or 28, wherein said antimicrobial composition contains at least one hydrophilic alcoholic GRAS flavoring agent and/or one hydrophilic non-alcoholic GRAS flavoring agent.

32. The method according to claim 31, wherein said antimicrobial composition further contains benzyl alcohol and/or a polyphenol compound (b).

33. The method according to claim 27 or 28, wherein said antimicrobial composition contains

(A) one or more GRAS flavor alcohols (a) or their derivatives; and

(B) one or more flavoring agents selected from

(b) polyphenol compounds; and

(c) GRAS flavor acids or their derivatives.

34. The method according to claim 33, wherein said antimicrobial composition contains

from 0.1 to 99% by weight, preferably from 0.5 to 99% by weight, of component (a),

from 0 to 25% by weight, preferably from 0.01 to 10% by weight, of component (b), and

from 0 to 70% by weight, preferably from 0.01 to 30% by weight, of component (c).

35. The method according to one or more of claims 27 to 34, wherein said antimicrobial composition contains benzyl alcohol and at least one further GRAS flavoring agent.

5 36. The method according to claim 35, wherein said further GRAS flavoring agents are selected from (a) GRAS flavor alcohols or their derivatives, (b) polyphenol compounds, (c) GRAS flavor acids or their derivatives, (d) phenols or their derivatives, (e) esters, (f) terpenes, (g) acetals, (h) aldehydes and (i) essential oils.

10 37. The method according to claim 36, wherein said antimicrobial composition contains less than 50% by weight, preferably less than 30% by weight, more preferably less than 20% by weight, of benzyl alcohol or of a mixture of benzyl alcohol with ethanol and/or isopropanol.

15 38. The method according to one or more of claims 33 to 36, wherein said antimicrobial composition contains

(a1) benzyl alcohol as a necessary component; and optionally

20 (a2) one or more further GRAS flavor alcohols or their derivatives; and

(b) one or more polyphenol compounds; and/or

(c) one or more GRAS acids or their derivatives.



39. The method according to claim 38, wherein said further GRAS flavor alcohol (a2) is selected from:

acetoin, ethyl alcohol, propyl alcohol, isopropyl alcohol, propylene glycol, glycerol, n-butyl alcohol, iso-butyl alcohol, hexyl alcohol, L-menthol, octyl alcohol, cinnamyl alcohol,  $\alpha$ -methylbenzyl alcohol, heptyl alcohol, n-amyl alcohol, iso-amyl alcohol, anisalcohol, citronellol, n-decyl alcohol, geraniol,  $\beta$ , $\gamma$ -hexenol, lauryl alcohol, linalool, nerolidol, nonadienol, nonyl alcohol, rhodinol, terpeneol, borneol, clineol, anisole, cuminyl alcohol, 10-undecene-1-ol, 1-hexadecanol or their derivatives;

said polyphenol compound (b) is selected from:

catechol, resorcinol, hydroquinone, phloroglucinol, pyrogallol, cyclohexane, resveratrol, usnic acid, acylpolyphenols, lignins, anthocyanins, flavones, catechols, gallic acid derivatives, caffeic acid, flavonoids, derivatives of the mentioned polyphenols, and extracts from Camellia, Primula; and

said GRAS acid (c) is selected from:

acetic acid, aconitic acid, adipic acid, formic acid, malic acid, capronic acid, hydrocinnamic acid, pelargonic acid, lactic acid, phenoxyacetic acid, phenylacetic acid, valeric acid, iso-valeric acid, cinnamic acid, citric acid, mandelic acid, tartaric acid, fumaric acid, and their derivatives.

40. The method according to claim 38 or 39, wherein said antimicrobial composition contains

from 0.1 to 99% by weight, preferably from 0.1 to 75% by weight, of benzyl alcohol;

from 0 to 99.8% by weight, preferably from 0.01 to 99% by weight, of component (a2); and

from 0 to 25% by weight, preferably from 0.01 to 10% by weight, of component (b);

from 0 to 70% by weight, preferably from 0.01 to 30% by weight, of component (c).

41. The method according to one or more of claims 38 to 40, wherein said antimicrobial composition contains further GRAS flavoring agents selected from (d) phenols, (e) esters, (f) terpenes, (g) acetals, (h) aldehydes and (i) essential oils.

42. The method according to claim 41, wherein said antimicrobial composition contains from 0.001 to 25% by weight, preferably from 0.01 to 9% by weight, of said further GRAS flavoring agents (d) to (i).

43. The method according to claim 41 or 42, wherein said further GRAS flavoring agents are phenols (d) and/or essential oils (i).

44. The method according to one or more of claims 27 to 43, wherein said antimicrobial composition does not contain any derivatives of the GRAS flavoring agents.

45. The method according to one or more of claims 38 to 44, wherein said antimicrobial composition contains one or two GRAS flavor alcohols (a2) and at least one polyphenol compound (b).

46. The method according to claim 45, wherein said polyphenol compound (b) is tannin.

47. The method according to claim 46, wherein said antimicrobial composition contains from 20 to 98% by weight of benzyl alcohol and from 0.01 to 10% by weight of tannin.

48. The method according to claim 27, wherein said antimicrobial composition is defined as in claims 1 to 25, especially as in claims 12 to 25.

49. A method for protecting plants shortly before or after the harvest from insects and insect larvae, comprising:

(i) the distribution or application of an insecticidal composition to the surface of the plants and/or

(ii) the distribution of an insecticidal composition within the plant by adding the insecticidal composition to nutrient media, nutrient liquids and/or water; wherein said insecticidal composition is a composition containing GRAS flavoring agents as defined in claims 1 to 25 and 27 to 48.

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50. The method according to claim 49, wherein said distribution or application of the insecticidal composition to the surface of the plant comprises application of said insecticidal composition to the surface of the plants, especially by spraying, immersion or nebulizing.

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51. The method according to one or more of claims 1 to 50, wherein said plants are selected from cotton, cereals, rice, corn, potatoes, tobacco, coffee, cocoa, tea, vegetables, fruits, nuts, spices, herbs, seeds, ornamental plants, cultured flowers and flowers for cutting.

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52. Use of an antimicrobial composition as defined in claims 1 to 25 and 27 to 48 for the treatment of plants shortly before and after the harvest.

**ABSTRACT**

The invention relates to methods for protecting plants and/or parts of plants from insects and insect larvae and from microbial attack. Said methods  
5 comprise the distribution or application of special insecticides or antimicrobial compositions on or to the surface of the plants and/or plant parts and/or the distribution of the compositions in the plant and/or the plant part. The invention also relates to insecticides and biocides containing these insecticides or antimicrobial compositions and to the use of the insecticides or antimicrobial  
10 compositions for treating plants and/or plant parts.

**DECLARATION, POWER OF ATTORNEY AND POWER TO INSPECT**

As the below named inventor, I hereby declare:

that my residence, post office address and citizenship are as stated below next to my name;

that I verily believe I am the original, first and sole inventor of the Invention entitled:

**"PLANT PROTECTION,"** United States Patent Application Serial No. 10/069476 filed February 25, 2002.

the specification of which [check one(s) applicable]:

- ☒ was first described and claimed in German Patent Application Serial No. **199 40 283.3** filed on **August 25, 1999**;  
☒ was filed as PCT International/U.S. Application No. **PCT/EP/00/08344** on **August 25, 2000** (International Filing Date)  
 and was amended by Amendments filed \_\_\_\_\_ (if applicable); [or];  
☒ is attached to this Declaration, Power of Attorney and Power to Inspect;

that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above; and

that I acknowledge my duty to disclose information which is material to the examination of this Application in accordance with Rule 56(a) [37CFR§1.56(a)].

**POWER OF ATTORNEY:** As inventor, I hereby appoint the practitioners associated with **Customer No. 000110** as my attorneys or agents with full power of substitution to prosecute this Application and to transact all business in the Patent and Trademark Office connected therewith: **John S. Child Jr., Reg. No. 28,833**

**POWER TO INSPECT:** I hereby give **DANN, DORFMAN, HERRELL AND SKILLMAN, P.C.** of Philadelphia, PA or its duly accredited representatives power to inspect and obtain copies of the papers on file relating to this Application.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the Application or any patent issued thereon.

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